



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Application of: Tsutomu FUKUDA et al.

Serial No.: 10/530,476

Group Art Unit: 1755

Filed: April 6, 2005

Examiner: KARL E. Group

For: METHOD FOR PRODUCING ALUMINUM MAGNESIUM TITANATE  
SINTERED PRODUCT

**DECLARATION**

Honorable Commissioner of Patents and Trademarks  
Washington, D.C. 20231

Sir :

I, Masaaki FUKUDA, hereby declare:

1) That I am one of the inventors of the instant invention, and

2) That the experiments given below were carried out under my general direction and supervision.

**Experiment 1**

**1. Purpose of Experiment**

The purpose of the experiment is to show the difference between a sintered body of aluminum magnesium titanate as defined in claim 5 of the present application and a sintered body of Example of Japanese document 4-280863.

**2. Sample preparation method**

(1) A sintered body of aluminum magnesium titanate was prepared following the procedure of Example 3 in the specification of this application. This sintered body is referred to as sample A.

(2) A sintered body of sample No. 3 shown in Fig. 4 of Japanese document 4-280863 was prepared following the procedure of Example of Japanese document 4-280863. The

mixing manner of raw materials, molding manner, and sintering conditions were the same as in sample A. This sintered body is referred to as sample B.

Table 1 shows the raw material composition of each sample.

Table 1

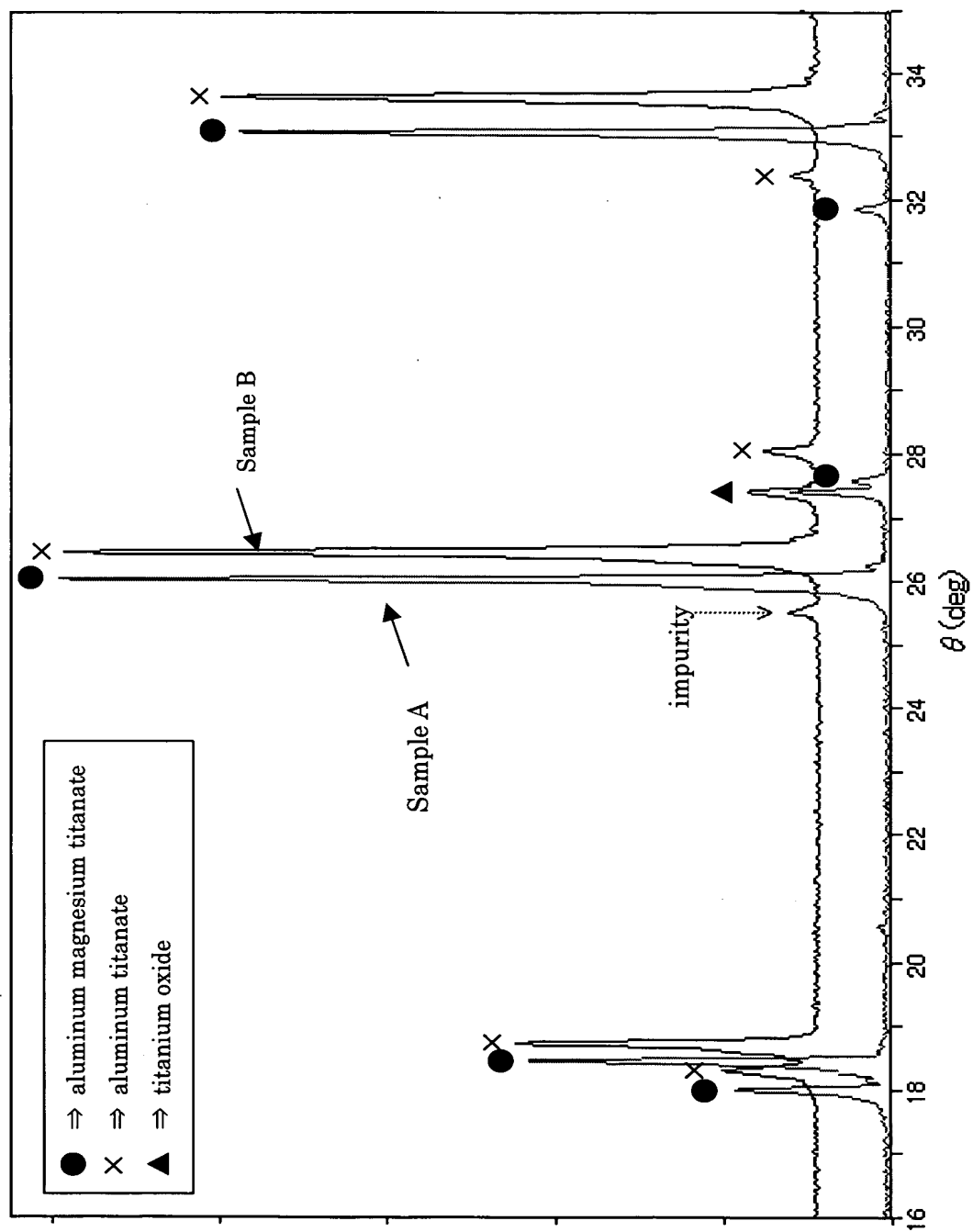
Sample	Raw material composition (part by weight)					
	Al <sub>2</sub> TiO <sub>5</sub>	Al <sub>2</sub> O <sub>3</sub>	TiO <sub>2</sub>	MgO	Alkali feldspar	Sumecton SA
A	-	43	52	5	4	-
B	90	-	5.0	-	-	5.0

### 3. Test method and test results

#### (i) Powder X-ray diffraction analysis:

The sintered body samples A and B above were analyzed in terms of the crystal structure by powder X-ray diffraction measurement (Device name: RINT2100, product of Rigaku Co., Ltd.). The sintered body samples were sufficiently crushed into powder in a mortar. A suitable amount of powder was placed in a sample folder, and then measured using an X-ray diffractometer. Fig. 1 illustrates X-ray diffraction spectra showing the crystal structures of samples A and B.

Fig. 1



(ii) Measurement of physical properties

The three-point bending strength of the sintered body samples A and B was determined following the procedure of Example 1 in the specification of the present application. The coefficient of thermal expansion of the samples was determined following the procedure of Example 2.

As a thermal-decomposition resistance test, sample A was evaluated for the change in residual aluminum magnesium titanate percentage  $\alpha(\%)$  over time following the procedure of Example 3 in the specification of the present application.

Separately, a 10 mm  $\times$  10 mm  $\times$  10 mm test sample was cut from sample B. The test sample was allowed to stand in air at 1100°C, and analyzed for the change in residual aluminum titanate percentage  $\beta(\%)$  over time according to the following method.

Measurement method of residual aluminum titanate percentage  $\beta(\%)$

The residual aluminum titanate percentage was determined from the X-ray diffraction (XRD) measurement spectrum by the following manner.

First, based on the fact that  $\text{Al}_2\text{O}_3$  (corundum) and  $\text{TiO}_2$  (rutile) are formed when aluminum titanate thermally decomposes, the ratio  $r$  of the diffraction intensity of aluminum titanate to the total diffraction intensity of aluminum titanate and rutile was determined by the formula below using the integrated intensity of the diffraction peak of the (110) face of rutile and the integrated intensity of the diffraction peak of the (023) face of aluminum titanate:

$$r = I_{\text{AT}(023)} / \{ I_{\text{AT}(023)} + I_{\text{TiO}_2(110)} \}$$

Further, the ratio  $r_0$  of the diffraction intensity of aluminum titanate to the total diffraction intensity of aluminum titanate and rutile for the sintered body prior to conducting heat treatment at 1100°C was also determined by the same manner.

Second, using  $r$  and  $r_0$  determined by the above manner, the residual aluminum titanate percentage  $\beta(\%)$  was determined by the following formula:

$$\beta = (r/r_0) \times 100$$

Table 2 shows, with respect to each sintered body, the three-point bending strength, coefficient of thermal expansion and residual aluminum magnesium titanate percentage  $\alpha(\%)$  or residual aluminum titanate percentage  $\beta(\%)$  after 300 hours had passed and 400 hours had passed in the thermal-decomposition resistance test.

Table 2

Sample	Three point bending strength (MPa)	Coefficient of thermal expansion ( $\times 10^{-6}/^{\circ}\text{C}$ )	Residual aluminum magnesium titanate or aluminum titanate percentage (%)	
			After 300 hours had passed	After 400 hours had passed
A	45.6	0.3	93.1	85.6
B	3.4	1.0	0.0	0.0

#### 4. Consideration

Comparison between the X-ray diffraction spectra of samples A and B shows that samples A and B are different in the diffraction angle of the diffraction peak of the main component as observed in X-ray diffraction spectra, and therefore the crystal structures are different from each other.

Moreover, as is clear from the physical property measurement, sample A has very much higher bending strength and much more excellent thermal decomposition resistance properties than those of sample B. Thus, it was confirmed that sintered body samples A and B were different in the crystal structure and the physical properties thereof were remarkably different from each other.

The above test results show that sample B, i.e., the sintered body of sample No. 3 of Example in Japanese document

4-280863, is different from the sintered body of aluminum magnesium titanate obtained in the present invention (sample A) .

## Experiment 2

### 1. Purpose of Experiment

The purpose of the experiment is to show the difference in thermal decomposition resistance between a sintered body of aluminum magnesium titanate obtained according to the method of the invention of the present application and sintered bodies according to the methods of Japanese document 2002145659 and Kameda (US 5,008,222).

### 2. Sample preparation method

(1) A sintered body of aluminum magnesium titanate was prepared in the same manner as in Example 4 in the specification of the present application. This sintered body is referred to as sample C-1.

Another sintered body of aluminum magnesium titanate was prepared in the same manner as in Example 4 in the specification of the present application except that the amount of alkali feldspar was 8 parts by weight, per 100 parts by weight of the total amount of  $\text{Al}_2\text{O}_3$ ,  $\text{TiO}_2$  and  $\text{MgO}$ . This sintered body is referred to as sample C-2.

Note that sintered body samples C-1 and C-2 were obtained according to the method of the present invention.

(2) Sintered bodies of aluminum titanate were prepared using a raw material mixture prepared by adding alkali feldspar in an amount of 4 parts by weight or 8 parts by weight to 100 parts by weight of a mixture of 56.1% by weight of  $\text{Al}_2\text{O}_3$  and 43.9% by weight of  $\text{TiO}_2$  in the same manner as in Example 4 in the specification of the present application. Used was alkali feldspar with the same composition as that of the alkali feldspar of sample C-1. The sintered body prepared by adding 4 parts by weight of the alkali feldspar is referred to as sample D-1, and the sintered body prepared by adding 8 parts by weight of the alkali feldspar is referred to as

sample D-2.

(3) Sintered bodies of aluminum titanate were prepared following the procedure of Example 4 in the specification of the present application using a raw material prepared by adding 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of a mixture of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  in the molar ratio of  $\text{Al}_2\text{O}_3:\text{TiO}_2=1.3:0.7$  or a raw material prepared by adding 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of the mixture. The sintered body prepared by adding 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-1-1, and the sintered body prepared by adding 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-1-2.

(4) Sintered bodies of aluminum titanate were prepared following the procedure of Example 4 in the specification of the present application using a raw material prepared by adding 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of a mixture of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  in the molar ratio of  $\text{Al}_2\text{O}_3:\text{TiO}_2=1.0:1.0$  or a raw material prepared by adding 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of the mixture. The sintered body prepared by adding 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-2-1, and the sintered body prepared by adding 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-2-2.

(5) Sintered bodies of aluminum titanate were prepared following the procedure of Example 4 in the specification of the present application using a raw material prepared by



adding 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of a mixture of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  in the molar ratio of  $\text{Al}_2\text{O}_3:\text{TiO}_2=0.8:1.2$  or a raw material mixture prepared by adding 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide to 100 parts by weight of the mixture. The sintered body prepared by the addition of 4 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-3-1, and the sintered body prepared by the addition of 8 parts by weight of magnesium oxide and 4 parts by weight of silicon oxide is referred to as sample E-3-2.

### 3. Test methods and Results

As a thermal-decomposition resistance test, the change in residual aluminum magnesium titanate percentage  $\alpha(\%)$  or residual aluminum titanate percentage  $\beta(\%)$  over time was determined for each sample left in air at a high temperature ( $1100^\circ\text{C}$ ) in the same manner as in Experiment 1 above.

Fig. 2 is a graph showing the change in residual percentage of each sintered body, and Table 3 summarizes the changes.

Fig. 2

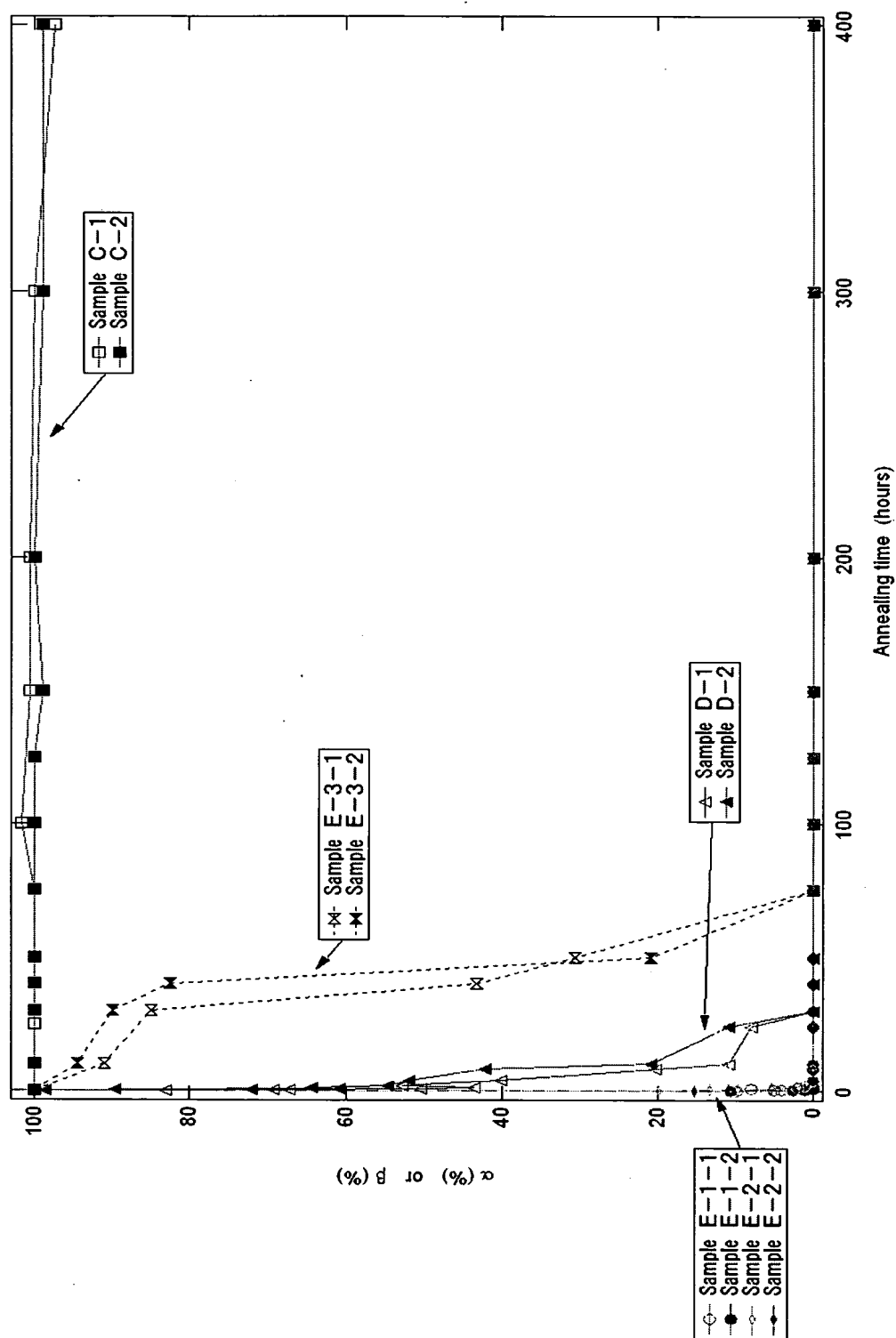


Table 3

Sample No.	Residual aluminum magnesium titanate percentage or residual aluminum titanate percentage (%)					
	10 hours after	30 hours after	50 hours after	100 hours after	200 hours after	400 hours after
C-1	100.0	100.0	100.0	101.7	100.7	97.4
C-2	100.0	100.0	100.0	100.0	100.0	98.9
D-1	10.8	0	0	0	0	0
D-2	20.9	0	0	0	0	0
E-1-1	0	0	0	0	0	0
E-1-2	0	0	0	0	0	0
E-2-1	0	0	0	0	0	0
E-2-2	0	0	0	0	0	0
E-3-1	91.1	85.0	30.7	0	0	0
E-3-2	94.6	90.1	20.9	0	0	0

#### 4. Consideration

(1) Sintered body samples C-1 and C-2 were obtained using the raw material mixture prepared by adding alkali feldspar to a raw material mixture for aluminum magnesium titanate, i.e., sintered bodies obtained according to the method of the present invention. As is clear from the thermal decomposition resistance tests for these sintered bodies, a sintered body obtained according to the method of the present invention can maintain a high value of the residual aluminum magnesium titanate percentage for a long period when left under a high temperature condition of 1100°C, and the sintered body has excellent resistance to thermal decomposition.

(2) Sintered body samples D-1 and D-2 were obtained using a raw material mixture prepared by adding alkali feldspar to a raw material mixture for aluminum titanate, i.e., sintered bodies obtained according to the method of Japanese document

2002145659. As revealed by the test results for these sintered bodies, sintered bodies obtained according to the method of Japanese document 2002145659 decompose in a short period, i.e., have poor thermal decomposition resistance at high temperatures. It was found that, with respect to the sintered body obtained using a raw material mixture prepared by adding alkali feldspar to a raw material mixture for aluminum titanate, the thermal decomposition resistance was merely slightly improved.

(3) Each of sintered body samples E-1-1 to E-3-2 was obtained using one of raw material mixtures prepared by adding magnesium oxide and silicon oxide to one of raw materials comprising  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  in a molar ratio of  $\text{Al}_2\text{O}_3:\text{TiO}_2 = 1.3:0.7$ ,  $1:1$  or  $0.8:1.2$ . These sintered bodies were obtained according to the method of Kamaeda (US 5,008,222). As is clear from the test results for these sintered bodies, the sintered bodies of Kamada (US 5,008,222) showed, depending on the raw material composition, slight improvement in the thermal decomposition resistance. However, compared with the sintered bodies of the present invention (samples C-1 and C-2), the sintered bodies of Kameda thermally decompose completely in an extremely short period, and thus, cannot be continuously used at high temperatures for long periods. Accordingly, it is also found in the case of the addition of magnesium oxide and silicon oxide to a mixture of  $\text{Al}_2\text{O}_3$  and  $\text{TiO}_2$  that the effect on improving thermal decomposition resistance of a sintered body is very slight.

\* \* \* \* \*

I, the undersigned, declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and

further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: June 22, 2006

Masaaki Fukuda

Masaaki FUKUDA